MULTISCALE MULTIPHYSICS MODEL FOR HYDROGEN EMBRITTLEMENT IN POLYCRYSTALLINE NICKEL

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Abstract

A multiscale multiphysics model has been developed to investigate hydrogen embrittlement in polycrystalline nickel. Appreciation of the role that hydrogen and residual stresses play in hydrogen embrittlement of polycrystalline metals has led the authors to propose a coupled microstructural and continuum approach to the simulation of the chemo-mechanical process. Within a macro scale finite element model the macro domain is replaced at critical defect sites with a microstructural domain. Resolution of the different microstructural material characteristics within the grain, grain boundary and triple junction points, including grain orientation can be used to improve the predicted distributions of both hydrogen and stress within the microstructure. The results show that microstructural features of polycrystalline nickel play an important role in the segregation of hydrogen which may lead to hydrogen induced crack initiation and development. Computational analyses are presented to demonstrate the application of the model in an attempt to bridge the gap between the micro and macro length scales for the chemo-mechanical problem in the case of a small, pre-placed nano-crack in a loaded plate in a hydrogen rich environment.

Keywords: Computer simulations, hydrogen absorbing materials, diffusion, grain boundaries, microstructure, mechanical properties.

1. Introduction:

Aerospace industry has experienced unexpected hydrogen embrittlement problems for many years and the structural failures due to hydrogen embrittlement of aerospace components made of polycrystalline face-centred cubic (FCC) nickel and nickel based super alloy are often sudden and unpredictable. They may occur after a few hours, a few days or sometimes it may take years after manufacturing the component using manufacturing process such as welding, electroplating, etc… [1-11]. Hydrogen embrittlement generally concerns interactions between

the microstructure, dislocations, hydrogen absorption/diffusion and stress [12-16]. Hydrogen and stress may be introduced into the metal during the manufacturing process. The chemical diffusion of hydrogen is inhomogeneous at the microstructural level in nickel due to the difference in the diffusivity within grains and at grain boundaries as reported by Harris *et al*. [12], Palumbo *et al*. [14] and Gertsman *et al*. [15]. The initial stress distribution in FCC polycrystalline nickel is often non-uniform and inhomogeneous at the microstructural level due to the random crystallographic texture and anisotropic properties of individual crystals, even under uniform applied stress. Inhomogeneous local stress distributions will impact on the stress assisted diffusion of hydrogen in polycrystalline nickel. They may play a critical role in the initiation of small micro cracks, such as intergranular cracks. The behaviour of these intergranular cracks is dependent on the inclination of the local grain boundary with respect to the tensile axis as reported in Kamaya *et al*. [17]. The growth of such cracks contributes to the potential failure of the material. It is therefore important to try to account for important microstructural phenomena within a component scale model to aid prediction of hydrogen embrittlement. In this work a multiscale coupled chemo-mechanical modelling technique in the form of stress assisted hydrogen diffusion is proposed. By accounting for (i) the effect of heterogeneous microstructures in critical sites and (ii) by coupling this with the homogeneous macro scale, one is able to investigate hydrogen embrittlement phenomena at the component level.

The proposed multiscale coupled Microstructure and Continuum Critical Defect model (MCCD) has been used to study the hydrogen embrittlement problem. The model solves the chemo-mechanical problem including local microstructural features in a selected critical defect site with boundary conditions interpolated from the solution from an initial macro global model. The critical defect micro-scale region contains microstructural details describing local heterogeneous microstructural features, in this case grain boundaries (GB), triple junctions (TJ) and individual grain orientations. Grain boundary regions are defined as critical microstructural units because both hydrogen diffusion and dislocations within the material will be significantly affected by these crystal defects. The mechanical and stress-assisted diffusion models are coupled sequentially. First the mechanical problem of the multiscale MCCD model is solved which is then used in the subsequent stress assisted hydrogen diffusion model. Thus the MCCD

model attempts to bridge the gap between microstructural and continuum length scales to investigate the chemo-mechanical hydrogen embrittlement problem.

2. Multiscale coupling of critical site microstructure and the continuum level – details of model and analysis:

2.1. Coupled MCCD model :

Figure 1 summarizes the length scales pertinent to this work. Figure 1 (a) shows the macro scale combustion chamber of rocket [25]. In particular, figure 1 (b) shows the geometry and dimensions of the 2D MCCD model containing the critical defect sites (grain boundaries) and its relationship with the macro scale homogeneous component in figure 1 (a). The MCCD model is developed using two modelling techniques, the first one using a "tie boundary technique" or "substructure technique" and the second one using a "cut boundary technique" or "submodelling technique". Detailed information about submodelling and substructuring techniques can be found elsewhere [19, 20]. The cut boundary technique often reduces analysis times when compared to the tie boundary technique. The procedure to couple a heterogeneous microstructural model to a homogeneous model using both tie boundary and cut boundary techniques is described by Jothi *et al*. [20].

2.2. The microstructural heterogeneous polycrystalline model:

Figure 1(c) shows the nano cracked microstructural model geometry and dimensions of the 2D heterogeneous microstructural polycrystalline model composed of polycrystalline aggregate grains and grain boundaries with an average grain size of 65 nm and average grain boundary affected zone thickness of 1 nm. Note the crack deliberately included at the bottom left of the grid. The crack dimensions (length=100nm and width=20nm) and load direction are also shown in figure 1(c). The full procedure to generate the microstructural model, i.e. the computational microstructure shown in figure 1 (c), can be found in Jothi *et al*. [26] It is summarised as follows:

a) Random points are generated in the required microstructural domain,
b) By keeping these random points, irregular non-overlapping polygonal grains are generated based on Voronoi tessellation theory,
c) Grain boundary affected zones of thickness 1 nm are added between grains,
d) Coordinate data points of microstructural features such as grains, grain boundary affected zones and TJs are meshed and grains are assigned individual orientations in ABAQUS using PYTHON scripting.

Figure 1(d) shows the close-up view of a single grain and its GB and grain boundary affected zone (GBAZ).

2.3.The Coupled Chemo-Mechanical Analysis with stress potential model:

The hydrogen embrittlement problem is solved as follows. The chosen problem in this paper is a cracked polycrystalline nickel plate subjected to loading in a hydrogen rich environment where a simulated crack is deliberately introduced into the mesh. The commercial finite element program ABAQUS was employed for a sequentially coupled stress-assisted hydrogen diffusion analysis consisting of repeating steps of a static stress analysis followed by a hydrogen diffusion analysis. A very fine mesh is used in the microstructural model to accurately capture the gradients of stress and hydrogen concentration near the artificial crack tip and a coarser mesh is used in the more distant (and assumed homogeneous) macro region. The procedure to couple the chemo-mechanical analysis is as follows:

a) The mechanical response of the coupled domain is captured firstly using an initial static stress analysis. Initially, the stress analysis is done at the macro-structural level, shown in figure 1 (b), assuming the material is isotropic with a modulus of elasticity of 200 GPa and Poisson's ratio of 0.3. These results are transferred to the microstructural level sub-mesh, shown in figure 1 (c), via the submodelling technique [19-20]. This effectively maps values from the macro model to the boundaries of the micro mesh. Values calculated in the micro region are similarly mapped back onto the macro mesh at the next stress analysis. The same macro to micro coupling procedure is carried out for the hydrogen diffusion analysis as well.

b) In the micro-scale model the material mechanical anisotropy is accounted for by assigning individual grains random crystallographic orientations using the FCC nickel single crystal elastic constants C_{11}=247 GPa, C_{12}=147 GPa and C_{44}=125 GPa [21,24].

c) The micro-level model also includes micro-level stress-based driving forces for hydrogen diffusion. Hydrogen diffusion without the effect of stress in an FCC single crystal is isotropic with one independent diffusion material property [22]. However, hydrogen diffusion in grain boundaries is faster than diffusion within grains in nickel. The hydrogen diffusivity and initial hydrogen concentration values used in the micro model are 3.52×10^{-10} cm^2/s (within grains) and 2.05×10^{-8} cm^2/s (in grain boundary affected zones) [24,26] and $1*10^{-4}$ mol.H/nm3 (i.e. initial hydrogen concentration) . Detailed descriptions and the equations used in the chemo-mechanical analysis are described in Jothi *et al*. [24]. Thus the model uses a relatively straightforward

coupling from macro to micro scales, assuming isotropic mechanical/diffusivity behaviour in the former and mechanical anisotropy and varying hydrogen diffusivity values in the latter.

3. Results and Discussion:

3.1. Stress distribution predictions from the MCCD model:

Figure 2(a) shows the von Mises stress distribution in the microstructural domain under a 1×10^{-11} N/nm^2 applied uniform load. The stresses observed near the defect are high as expected. High stresses near the crack tips are heterogeneous with high stresses near and along the grain boundary. The stresses in the rest of the microstructural region are heterogeneous between grains and grains boundaries for the uniform applied stress. These heterogeneous stress distributions show high stresses near the grain boundaries and triple junctions when compared to grain interiors due to the anisotropy arising from the random crystallographic orientation constraints and deformation constraints caused by the grains and adjacent neighbouring grains as reported in the previous research [17,23,24,27]. Figure 2(b) shows the shear stress in the microstructural domain.

High shear values are observed near and along GBs and TJs (highlighted in dotted circles) and also shear gradients are typically higher between neighbouring grains. Stress analysis has also been done using isotropic material parameters (i.e. no crystal orientation effects). The results, contained in Figure 2(c), show a homogeneous stress distribution across grains and GBs as expected. It should be noted that stresses in the vicinity of the crack tip are higher when microstructural inhomogeneity is included (i.e. S^m=65.1721E-012 N/nm^2), Figure 2 (a) and lower for the homogeneous case (i.e. S^I=53.4389E-012 N/nm^2) in Figure 2 (c).

3.2. Hydrogen distributions arising from the chemo-mechanical response of nickel in the MCCD model:

Figure 3(a) shows the multiphysics chemo-mechanical response in the form of a hydrogen distribution with associated hydrogen accumulation in the microstructural region ahead of the defect tip after few seconds of elapsed time. It also shows significant hydrogen accumulation along GBs and TJs due to high tensile stresses induced in the microstructure. The rapid diffusion paths along GBs and TJs combined with high tensile stresses leads to accumulations of hydrogen

atoms forming clusters of high hydrogen concentration. Lower hydrogen accumulation is observed along GBs and TJs near regions of compressive stress. Compressive stresses coupled with faster diffusion along GBs and TJs effectively pushes the hydrogen atoms out from GB and TJ zones. Figure 3(b) and 3(c) shows the chemo-mechanical response in the micro domain without the mesh and with the mesh respectively, Figure 3(d) shows the mechanical response as well chemo-mechanical response along the normalized distance in the micro domain along the arrow line shown in Figure 3(c). "G" represents grain and "GB" represents GB in this figure. The dotted line curve shows the stress concentration in N/nm^2 and the solid line curve shows the hydrogen concentration in $mol.H/nm^3$.

Changes in local stress and hydrogen concentration at the grain scale are observed when moving from one grain to another grain. At GBs a peak value is often observed. The triple junctions display high accumulations of hydrogen compared to grains and other grain boundaries. These effects are due to neighbouring grain shapes and orientations. TJs are a meeting point of three or more GBs. Figure 3(e) shows the variation in hydrogen concentration with distance moving along various GBs towards various TJs. There are 130 data points along 18 GBs leading to 5 TJs plotted in this figure. This shows that when moving towards a TJ along a GB the predicted hydrogen concentration values (i) increase moving towards some TJs and (ii) decrease moving towards other TJs. Whether a high or low accumulation of hydrogen at triple junctions occurs is dependent on the tensile or compressive stress respectively in addition with the orientation of the grains and neighbouring grains as well as grain shape in the vicinity of the TJ. The prediction of tensile or compression stress at GBs and TJs depends on the orientation of grains and their neighbouring grains as well as grain shape in the vicinity of the TJ. The results here show it is important to consider the microstructural features of the material to better capture local stresses and the accumulation of hydrogen when investigating hydrogen embrittlement in polycrystalline materials.

4. Conclusion:

In order to better solve the multiscale coupled multiphysics chemo-mechanical hydrogen embrittlement problem of aerospace components made of polycrystalline nickel, a multiscale coupled microstructure and continuum critical defect method is proposed accounting for microstructural features within a macro scale homogeneous modelling framework. In this work a

stress-assisted hydrogen diffusion model was developed within ABAQUS employing the finite element method. The results from the model can be summarised:

- Stresses developed in the polycrystalline nickel microstructure are inhomogeneous, even though the applied stress is uniform. In the model this is due to the anisotropic mechanical response at the microstructural or grain level.
- Stresses in the grain boundary affected zone can be significant depending on the relative orientation of neighbouring grains.
- Different levels of hydrogen accumulation at grain boundaries and triple junctions are also predicted by the model. The orientation of neighbouring grains and grain boundary angles relative to the applied load gives rise to either tensile or compressive local stresses. It is these local tensile or compressive stresses that determine whether higher or lower local hydrogen accumulation occurs. This highlights the importance of such microstructural features in hydrogen embrittlement.
- Hydrogen accumulation in the microstructure is heterogeneous due to the inhomogeneous microstructural stress distribution coupled to varying hydrogen diffusivity at different regions in the microstructure.
- The MCCD model appears promising for the systematic investigation of hydrogen embrittlement scenarios in polycrystalline nickel accounting for important microstructural features such grains, grain boundaries and grain orientations. Clearly the next stage of model development will be to develop a full 3D version of the MCCD code.

Acknowledgements

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References:

[1] S.P. Lynch, Failures of engineering components due to environmentally assisted cracking, Practical Failure Analysis 3 (2003), pp.33-42.

[2] D.P. Williams, H.G. Nelson, Gaseous hydrogen embrittlement of Aerospace materials, Astronautical Research (1973), pp.323-330.

[3] V. Frick, G.R. Janser and J.A. Brown, in Space Shuttle Materials, Vol. 3, Society of Aerospace Materials and Process Engineers (1971), pp.597-634.

[4] Eggert D.Reese, Wolfgang Von Bestenbostel, Torsten Sebald, Georgios Paronis, Diego Vanelli and Yves Muller, Hydrogen embrittlement of pulse-plated nickel, JOM 66 (2014) pp.1368-1376.

[5] R.P. Jewet, R.J. Walter, W.T. Chandler and R.P. Frohmberg, Hydrogen-Environment Embrittlement of Metals, NASA Technology Survey (1972).

[6] Eggert D.Reese, "EU FP7 Multiscale modelling hydrogen Embrittlement- MultiHy" report, Airbus Group Innovations report (2012).

[7] Torsten Sebald, "EU FP7 Multiscale modelling hydrogen Embrittlement- MultiHy" report, Airbus Defense and Spaces report (2013).

[8] S. Jothi, T.N. Croft, S.G.R. Brown, "EU FP7 Multiscale modelling hydrogen Embrittlement- MultiHy" report, Swansea University report (2014).

[9] S.J. Klima, A.J. Nachtigall, C.A. Hoffman, Preliminary investigation of effect of hydrogen on stress-rupture and fatigue properties of an Iron-, a Nickel-, and a Cobalt-base Alloy, NASA Report TN D-1458, National Aeronautics and Space Administration, Cleveland, Ohio, Dec. 1962.

[10] Hugh R. Gray, Hydrogen Environment Embrittlement, Technical paper report (1972).

[11] J. Woodtli, R.Kieselbach, Damage due to hydrogen embrittlement and stress corrosion cracking, Engineering Failure Analysis, 7 (2000) pp.427-450.

[12] T.M. Harris, R.M. Latanision, Grain boundary diffusion of hydrogen in nickel, Metallurgical Transactions A, 22A (1991) pp. 351-355.

[13] T. Tsuru, R.M. Latanision, Grain boundary transport of hydrogen in nickel, Scripta Metall 16 (1982) pp. 575-578.

[14] G. Palumbo, and D.M. Doyle, and A.M. El-Sherik, and , U. Erb and K.T. Aust, , Intercrystalline hydrogen diffusion transport in nanocrystalline nickel, *Scripta Metall. Mater.*, **Vol. 25**, 1991, pp. 679–684.

[15] V.Y. Gertsman, , M. Janecek, and K. Tangri, , *Acta Mater.*, **Vol. 44**, 1996, pp. 2869–2882.

[16] A. Oudriss, J. Creus, J. Bouhattate, E. Conforto, C. Berziou, C. Savall, X. Feaugas, Grain size and grain boundary effects on diffusion and trapping of hydrogen in pure nickel, Acta Materialia 60 (2012) pp. 6814-6828.

[17] M. Kamaya, A simulation on growth of multiple small cracks under stress corrosion, Int. J. Fract., 130 (2004) pp. 787-801.

[18] S. Jothi, T.N. Croft, S.G.R. Brown, E. de Souza Neto, Finite Element Microstructural Homogenization techniques and Intergranular, intragranular microstructural effects on effective diffusion coefficient of heterogeneous polycrystalline composite media., Composite Structures,108 (2014) pp. 555-564.

[19] ABAQUS Version 6.11. User's manual, Dassault Systemes, 2011.

[20] S. Jothi, T.N. Croft, S.G.R. Brown, E.de Souza Neto, Multiscale modelling of Hydrogen embrittlement in polycrystalline material, *in 12th International conference on structural Integrity Assessment*. May 2013, Manchester, UK.

[21] J.F. Nye, Physical properties of Crystals, (1985), Oxford: Clarendon

[22] H. Mehrer, Diffusion in solids: Fundamentals, Methods, Materials, Diffusion-Controlled processes, (2007), Springer.

[23] T. Sumigawa, T. Kitamura, K. Ohishi, Slip behaviour near a grain boundary in high-cycle fatigue of poly-crystal copper, Fract. Eng. Mater. Struct. 27 (2004) pp. 495-503.

[24] S. Jothi, T.N. Croft, S.G.R. Brown, Influence of grain boundary misorientation on hydrogen embrittlement in bi-crystal nickel, Journal of hydrogen energy, (2014) (In Press), http://dx.doi.org/10.1016/j.ijhydene.2014.07.020.

[25] http://cs.astrium.eads.net/sp/launcher-propulsion/rocket-engines/vulcain-rocket-engine.html (last accessed 15/October/2013).

[26] S. Jothi, T.N. Croft, S.G.R. Brown, Mesoscale microstructural computational simulation of hydrogen permeation test to calculate the intergranular/grain boundary diffusivity, MH2014 Symposium Proceeding, July (2014) Manchester, UK.

[27] S. Jothi, T.N. Croft, S.G.R. Brown, Analysis of the effect of grain boundaries and trapping on the effective diffusivity of hydrogen in polycrystalline nickel, ICHP2014 conference proceeding, Feb(2014) Fukuoka, Japan.

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All Figures:

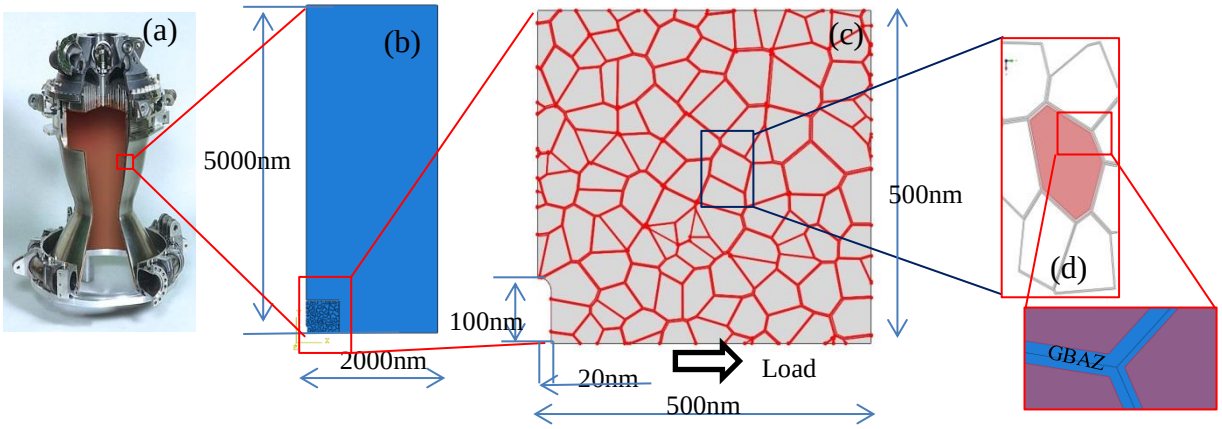

Figure 1. (a) Shows the macro scale combustion chamber of a rocket [14]. (b) Geometry of 2D MCCD macro model domain. (c) Microstructural polycrystalline model composed of grains and grain boundary affected zones (darker) with a pre-placed nano crack at bottom left. (d) Close-up view of nano grain (shaded), grain boundaries and below a higher magnification showing grain boundary affected zones (GBAZ).

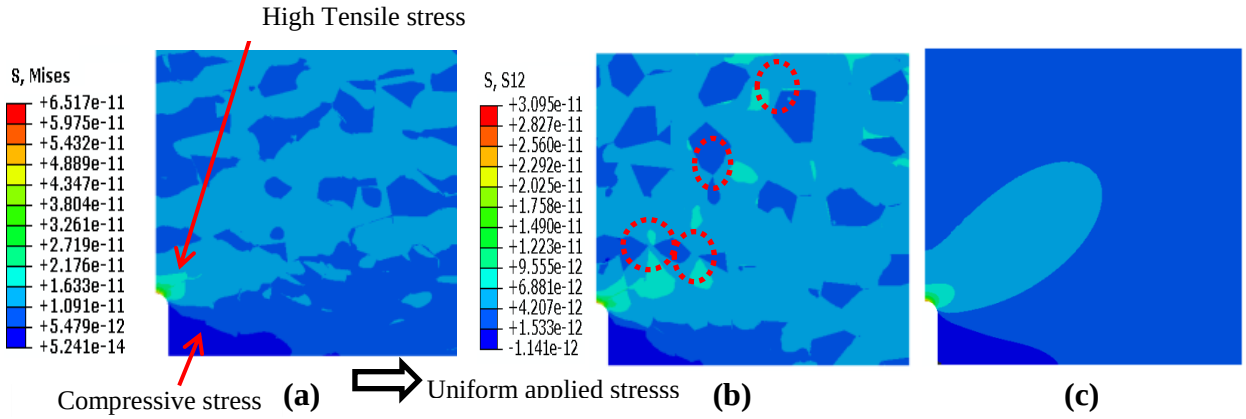

Figure 2. Shows the mechanical response in the microstructural region for uniform applied stress (a) von Mises stress (N/nm^2), (b) Shear stress in (N/nm^2), (c) shows the predicted von Mises stress (N/nm^2) without microstructural anisotropy effects.

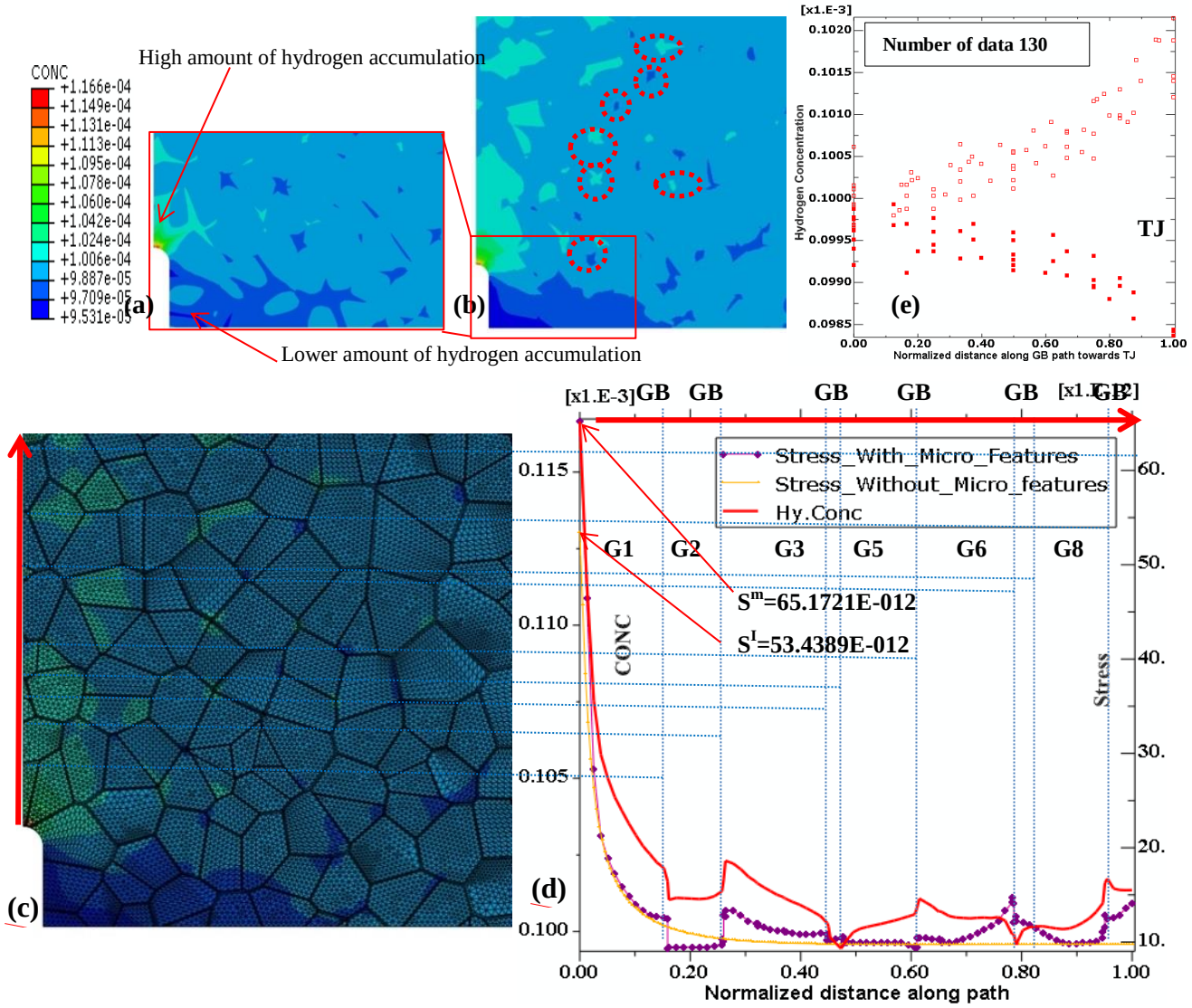

Figure 3. Shows the redistribution of hydrogen in the microstructural domain after a few seconds, (b) and (c) hydrogen distributions without mesh and with mesh respectively (Dotted circles show the accumulation of hydrogen on triple junctions, (d) hydrogen distribution (solid line and its unit in mol.H/nm^3) and stress distribution (dotted line) along the normalized path along arrowed line shown in Figure 3(c). It also shows the stresses in the vicinity of the crack tip are higher in model with microstructural feature (i.e S^m=65.1721E-012 N/nm^2) and lower in model without microstructure feature(i.e S^I=53.4389E-012 N/nm^2). (e) Shows the change in hydrogen concentration with the normalized distance along various GB paths towards TJs (i.e the value 1 in x-axis is the TJ). The hydrogen concentration values for 130 data are collected along 18 GBs belong to 5TJ s (i.e among 5TJ, 3TJ have four GBs per TJ and 2TJ have 3GB per TJ).